

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

URS LEO STADLER

APPLICATION NO: 10/762,601

FILED: JANUARY 22, 2004

FOR: STABILIZER COMPOSITION FOR THE  
ROTOMOLDING PROCESS

Group Art Unit: 1713

Examiner: Peter D. Mulcahy

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

**DECLARATION UNDER RULE 132**

James H. Botkin , the undersigned, states:

That I received a Ph.D. in Chemistry from Northeastern University, Boston, Massachussets, 1986;

That I have been employed by Ciba Specialty Chemicals Corporation and its predecessor corporation since 1989 ;

That I have approximately 24 years of chemical research and development experience; that from October 1989 to date I have worked in the Research and Development laboratories of Ciba Specialty Chemicals Corporation;

That the following experiments were performed by me or under my supervision:

Linear low density polyethylene powder (hexene copolymer, nominal melt index 3.3 dg/min, density 0.938 g/cm<sup>3</sup>) is dry blended with zinc stearate (0.05% by weight) and additional additives (see below). The mixtures are melt compounded into pellets using a Superior/MPM single screw extruder (24:1 L/D). The compounded pellets are ground to a uniform particle size powder using a Reduction Engineering Model 50 pulverizer with a gap size of 0.330 mm and at a temperature of 60-70°C.

Rotational molding is conducted using a FSP M20 clamshell-type molding machine equipped with a cubic (12" sides) cast aluminum mold. The mold is charged with 2020g ground polymer powder and heated at an oven temperature of 274°C (525°F) with biaxial rotation. Heating is maintained for a specified time (see Tables below). Subsequently, the oven is opened and while still rotating, the mold is cooled with forced air circulation for 7 minutes, followed by water spray mist for 7 minutes, and an additional air cooling step for 2 minutes. Throughout the entire molding cycle the speed of the primary axis of rotation is maintained at 6 rpm and the speed of the secondary axis rotation is maintained at 1.5 rpm. After the cooling is completed the mold is opened and the hollow object removed.

In Formulation 1 the polymer is additionally blended with a combination of the phosphite processing stabilizer IRGAFOS® 168 (0.12% by weight), the amine N-oxide GENOX® EP (0.05% by weight), and the hindered amine light stabilizer TINUVIN® 783 (0.20% by weight).

In Comparative Formulation 1 the polymer is additionally blended with a combination of the phosphite processing stabilizer IRGAFOS® 168 (0.12% by weight), the hindered phenolic antioxidant IRGANOX® 1010 (0.05% by weight), and the hindered amine light stabilizer TINUVIN® 783 (0.20% by weight).

In Comparative Formulation 2 the polymer is additionally blended with a combination of the phosphite processing stabilizer IRGAFOS® 168 (0.12% by weight), the hindered phenolic antioxidant IRGANOX® 3114 (0.035% by weight), and the hindered amine light stabilizer TINUVIN® 783 (0.20% by weight).

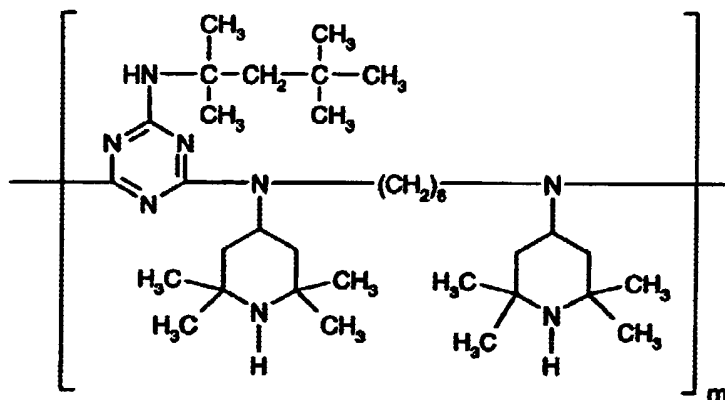
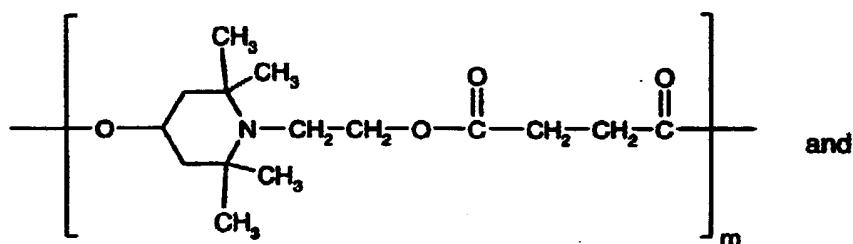
IRGAFOS® 168 is tris-(2,4-di-tert-butylphenyl)phosphite.

GENOX® EP is a di(C<sub>16</sub>-C<sub>18</sub>)alkyl methyl amine oxide, CAS# 204933-93-7.

**IRGANOX® 1010 is pentaerythritol tetrakis [3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate].**

**IRGANOX® 3114 is tris(3,5-di-tert-butyl-4-hydroxybenzyl) isocyanurate.**

**TINUVIN® 783 is a combination of hindered amines of formulae**



Each formulation is rotationally molded into hollow articles according to the general procedure with hold times ranging from 10 to 17 minutes.

The Yellowness Index of the inner surface of the hollow parts is determined using a DCI SF600 spectrophotometer according to ASTM D 1925. An increase in yellowness corresponds to a positive increase in Yellowness Index. Formulation 1 is superior to the Comparative Formulation 1 with regard to color stability after rotational molding at longer heating times ( $\geq 14$  minutes) and superior to the Comparative Formulation 2 over the entire range of heating times. The results are summarized in Table 1.

**TABLE 1**

<b>Heating Time at 274°C (min)</b>	<b>Yellowness Index</b>		
	<b>Formulation 1</b>	<b>Comparative Formulation 1</b>	<b>Comparative Formulation 2</b>
<b>10</b>	<b>-1.9</b>	<b>-1.2</b>	<b>0.3</b>
<b>12</b>	<b>-0.6</b>	<b>-0.7</b>	<b>4.4</b>
<b>13</b>	<b>1.7</b>	<b>0.4</b>	<b>14.6</b>
<b>13.5</b>	<b>3.0</b>	<b>3.6</b>	<b>20.1</b>
<b>14</b>	<b>3.4</b>	<b>5.2</b>	<b>27.2</b>
<b>15</b>	<b>2.1</b>	<b>11.4</b>	<b>25.8</b>
<b>15.5</b>	<b>2.3</b>	<b>14.3</b>	<b>19.2</b>
<b>17</b>	<b>14.8</b>	<b>21.2</b>	<b>30.4</b>

Low-temperature impact strength testing is performed using an instrumented drop weight impact apparatus DYNATUP® model 8250 according to ASTM D 3763. Test specimens cut from the rotomolded boxes are conditioned in an air circulated freezer for not less than 48 hours at -40°C prior to testing. The outside surface of the test specimens is subjected to the impact.

The impact results, reported as total impact energy, are summarized in Table 2. Formulation 1 proves to be superior to the Comparative Formulation 1 with regard to the low temperature impact strength, in particular after rotational molding with heating times <14 minutes. Formulation 1 is also superior to the Comparative Formulation 2 with regard to low temperature impact strength, in particular after rotational molding with heating times of <13 minutes.

**TABLE 2**

Heating Time at 274°C (min)	Total Impact Energy (ft-lb)		
	Formulation 1	Comparative Formulation 1	Comparative Formulation 2
10	23.5	14.7	19.7
12	27.6	19.2	21.4
13	33.1	19.0	30.3
13.5	32.9	26.6	33.5
14	33.2	31.5	33.0
15	33.2	32.1	33.7
15.5	32.4	33.9	30.9
17	10.9	15.3	14.0

In order to maximize molding productivity and reduce energy consumption it is important to be able to mold high-impact parts with a minimum heating time. Parts should also exhibit a minimum of discoloration. Table 3 summarizes the heating time required to attain optimum impact strength and the Yellowness Index after that heating time. Formulation 1 is superior to the Comparative Formulations in that it requires the shortest heating time to achieve optimum impact strength and exhibits the lowest Yellowness Index for parts with optimum impact strength.

**TABLE 3**

Formulation	Heating Time (min at 274°C) to Optimum Impact Strength	Total Impact Energy (ft-lb)	Yellowness Index
1	13	33.1	1.7
Comparative 1	15	32.1	11.4
Comparative 2	13.5	33.5	20.1

The present stabilizer system, Formulation 1, is clearly superior to two state-of-the-art stabilizer systems, Comparative Formulations 1 and 2, each of which contain hindered phenolic stabilizers in place of the present amine oxide stabilizer.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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James H. Botkin